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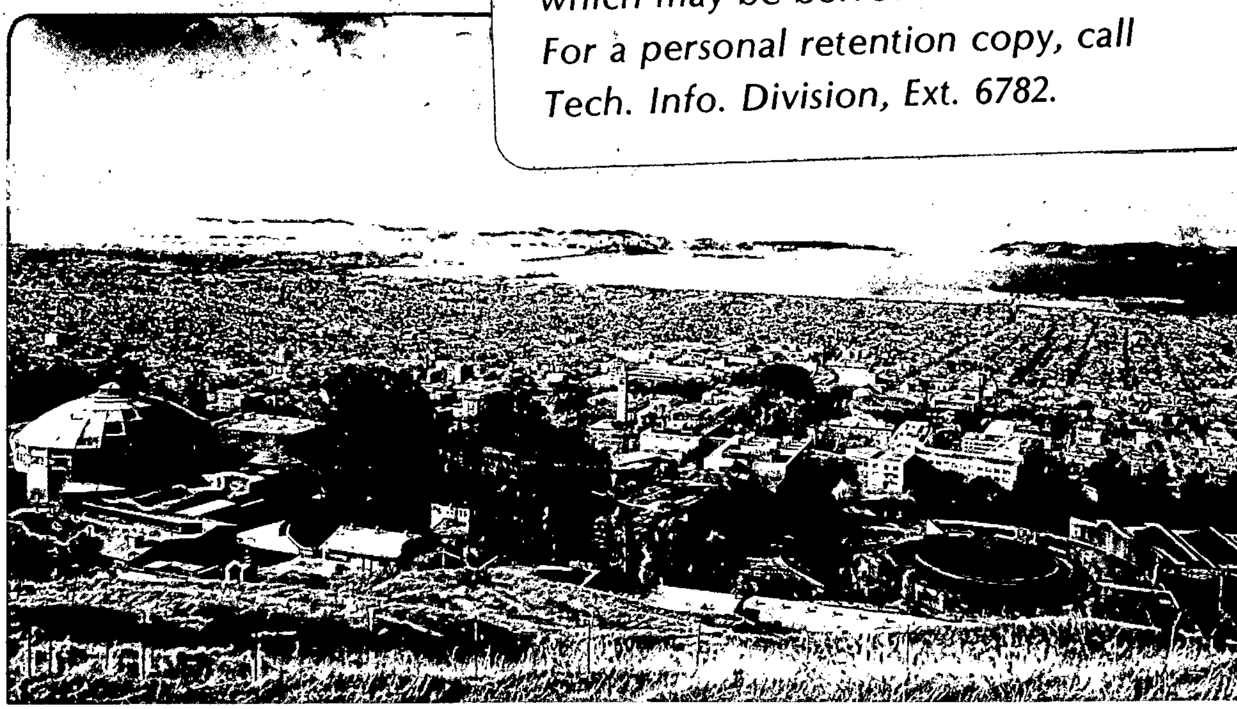

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INERTIAL EFFECTS IN TRANSPORT OF RADIOACTIVE WASTE THROUGH ENGINEERED BARRIERS

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ABSTRACT

Addition of an inertial term to the constitutive relation for mass flux leads to a mass transport equation which is hyperbolic and describes propagation of a distorted wave with a finite velocity. This approach eliminates the "instantaneous propagation paradox" inherent in parabolic transport equations based on Fick's law. Analytical solutions of the wave transport equation have been derived and have the properties that the leading edge of a solute front propagates at a finite, predictable velocity and is truncated by a step function which decreases in magnitude exponentially with time. The inertial effects on computed solute fronts are most evident near the leading edge, and have potential significance in the prediction of engineered barrier performance.

INTRODUCTION

Mathematical models are being used increasingly to predict movement of radioactive waste through engineered barriers and to interpret results of transport experiments with barrier materials. Model predictions of transport in engineered barriers may be used as input to models of transport in the far field. Because these models will be used as indicators of engineered barrier performance, and eventually in support of license applications, it is important that the models be as physically realistic as possible.

Let us examine the conceptual basis of many current models, and its consequences.

TRANSPORT EQUATION WITHOUT INERTIA

Mathematical models used to predict the performance of proposed engineered barrier materials traditionally have been constructed by combining the mass conservation equation with a constitutive relation describing the dependence of mass flux on forces acting on the system. For the simplest kind of transport model describing diffusion in one dimension, the mass conservation equation has the form

$$\frac{\partial C}{\partial t} + \frac{\partial j}{\partial x} = 0 \tag{1}$$

where C is the concentration of diffusing material, j is the diffusive mass flux, x is distance from the origin, and t is time.

Constitutive relations have taken a variety of forms; the usual practice in mass transport is to ignore thermodynamic couplings to other transport processes, to approximate chemical potential gradients by concentration gradients, and to write the constitutive relation for a single diffusing species in the form of Fick's law. In this form, the diffusive mass flux is stated to be proportional to the negative gradient of concentration through a transport coefficient, D, which in a fluid-saturated porous matrix is the coefficient of hydrodynamic dispersion. Thus,

$$j = -D\frac{\partial C}{\partial x}. \tag{2}$$

Combination of (1) and (2) results in a parabolic partial differential equation of transport, the "diffusion equation", which in this case has the form

$$D\frac{\partial^2 C}{\partial x^2} - \frac{\partial C}{\partial t} = 0. \tag{3}$$

When (3) is solved with the initial and boundary conditions

$$C(x,0) = 0, \qquad C(0,t) = C_0, \qquad \lim_{x\to\infty} C(x,t) = 0 \tag{4}$$

the result is

$$C(x,t) = C_0 \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right) \tag{5}$$

where $\operatorname{erfc}(z)$ represents the complementary error function with argument z. This procedure is subject to criticism on two grounds.

First, the simple phenomenological relation embodied in Fick's law was deduced from consideration of systems in steady states, and does not account for transient, inertial effects. Its application to systems showing explicit temporal dependence is questionable.

Second, solutions of the diffusion equation have the property that a material or thermal anomaly must be propagated with infinite velocity, i.e., the velocity of a concentration isopleth becomes large without bound as either time or concentration approaches zero. This can be shown for the present example by explicit derivation of the velocity of propagation, v_C, of an isopleth of given concentration C:

$$v_C = \left.\frac{dx}{dt}\right|_C = -\frac{\partial C/\partial t}{\partial C/\partial x} = \frac{x}{2t}. \tag{6}$$

Examination of (5) and (6) shows that v_C goes to infinity as either t or C approaches zero. This non-physical result (the "instantaneous propagation paradox") has received attention in recent years in the literature of heat transport [1,2,3,4]. We are led to inquire how we can avoid this paradox while retaining features of our transport models that correspond to our observational experience.

The paradox can be resolved by careful consideration of inertial processes (i.e., temporal changes of forces and fluxes) in a transporting system. Thus, Bearman and Kirkwood [5] and Bearman [6,7] used a statistical mechanical analysis of momentum balance, and Machlup and Onsager [8], Luikov [9], Gyarmati [10] and others used the thermodynamics of irreversible processes to derive constitutive relations including inertial terms explicitly.

The simplest form of an inertial term is the negative time derivative of a diffusive flux multiplied by a characteristic relaxation time, τ; this term is added to the diffusive term in Fick's law to give the complete constitutive relation for a given flux; thus,

$$j = -D\frac{\partial C}{\partial x} - \tau\frac{\partial j}{\partial t} \tag{7}$$

The result of combining such a relation with the mass conservation equation is a hyperbolic partial differential equation of transport describing a propagated wave with distortion caused by dissipative processes.

For example, the mass conservation equation for advective-dispersive transport in a one-dimensional system with fluid velocity equal to v is

$$\frac{\partial C}{\partial t} + v\frac{\partial C}{\partial x} + \frac{\partial j}{\partial x} = 0. \tag{8}$$

Combining (7) and (8) gives the hyperbolic partial differential equation

$$D\frac{\partial^2 Y}{\partial x^2} - v\tau\frac{\partial^2 Y}{\partial x \partial t} - \tau\frac{\partial^2 Y}{\partial t^2} - \frac{\partial Y}{\partial t} - v\frac{\partial Y}{\partial x} = 0, \tag{9}$$

where

$$Y(x,t) = C(x,t) \quad \text{or} \quad j(x,t).$$

To solve (9), we impose the initial conditions

$$C(x,0) = 0, \quad j(x,0) = 0, \quad \frac{\partial C(x,0)}{\partial t} = 0, \quad \frac{\partial j(x,0)}{\partial t} = 0. \tag{10}$$

The inner boundary condition can specify either a constant concentration, C_0,

$$C(0,t) = C_0, \tag{11}$$

or a constant flux, J_0,

$$vC(0,t) + j(0,t) = J_0. \tag{12}$$

In either case, the outer boundary conditions are:

$$\lim_{x\to\infty} C(x,t) = 0, \quad \lim_{x\to\infty} j(x,t) = 0. \tag{13}$$

The solutions of (9) to (13) have been obtained by an integral transform method. It is convenient to express the solutions in terms of the following reduced variables:

$$\xi = \frac{vx}{D}, \qquad \eta = \frac{v^2 t}{D}, \qquad \beta = \frac{v^2 \tau}{D},$$

$$\Delta = \frac{1}{2}\sqrt{\beta+4} - \frac{1}{2}\sqrt{\beta}, \qquad \eta_w = \Delta\xi\sqrt{\beta},$$

$$r_1 = \frac{\Delta^2}{\beta(\beta+4)}(\eta + \eta_w + \beta\xi), \qquad s_1 = \frac{\eta - \eta_w}{\beta(\beta+4)\Delta^2},$$

$$r_2 = \frac{r_1}{\Delta^4}, \qquad s_2 = \Delta^4 s_1,$$

$$z = 2\sqrt{r_1 s_1} = 2\sqrt{r_2 s_2}.$$

For the constant-concentration boundary condition, the solutions are:

$$\frac{C}{C_0} = \{J(r_1, s_1) + [1 - J(s_2, r_2)]\exp(\xi)\}H(\eta - \eta_w), \tag{14}$$

$$\frac{j}{vC_0} = \left\{\frac{I_0(z)}{\Delta\sqrt{\beta}}\exp\left[-\frac{(\beta+2)\eta}{\beta(\beta+4)} + \frac{\xi}{\beta+4}\right] - J(r_2, s_2)\exp(\xi)\right\}H(\eta - \eta_w), \tag{15}$$

where

$$J(r, s) = \exp(-s)\int_r^\infty \exp(-u)\, I_0(2\sqrt{su})\, du, \tag{16}$$

$I_n(z)$ is a modified Bessel function of order n with argument z, and $H(\eta)$ is the unit step function defined by:

$$H(\eta) = \begin{cases} 0 & \text{for } \eta < 0 \\ 1 & \text{for } \eta \geq 0 \end{cases}. \tag{17}$$

For the constant-flux boundary condition, the solutions are:

$$\begin{aligned} \frac{vC}{J_0} = H(\eta - \eta_w)\Big\{ & J(r_1, s_1) - g_1(\xi, \eta)J(r_2, s_2)\exp(\xi) \\ & + \left[g_2(\xi, \eta)I_0(z) + \frac{1}{2}\sqrt{\beta(\beta+4)}\, z I_1(z)\right]\exp\left[-\frac{(\beta+2)\eta}{\beta(\beta+4)} + \frac{\xi}{\beta+4}\right]\Big\}, \end{aligned} \tag{18}$$

$$\frac{j}{J_0} = H(\eta - \eta_w)\Big\{[1 + g_1(\xi,\eta)]J(r_2, s_2)\exp(\xi)$$
$$-\Big[[1 + g_2(\xi,\eta)]I_0(z) + \frac{1}{2}\sqrt{\beta(\beta+4)}zI_1(z)\Big]\exp\Big[-\frac{(\beta+2)\eta}{\beta(\beta+4)} + \frac{\xi}{\beta+4}\Big]\Big\}, \tag{19}$$

where

$$g_1(\xi,\eta) = 1 + \beta + (1+\beta)\xi + \eta, \qquad g_2(\xi,\eta) = \frac{\sqrt{\beta}}{\Delta} + \frac{\Delta\eta + \xi\sqrt{\beta}}{\Delta^3\sqrt{\beta(\beta+4)}}. \tag{20}$$

In these solutions, the variable η_w represents the time when the solute wave front reaches an observer located at a position represented by ξ; during the time period represented by $\eta < \eta_w$ the observer sees nothing. Thus the solute front moves at a constant, finite velocity; it can be shown that this propagation velocity, v_w, is given by:

$$v_w = \frac{2v}{\sqrt{\beta}\left(\sqrt{\beta+4} - \sqrt{\beta}\right)}. \tag{21}$$

Numerical values of the solutions given by (14) to (20) have been computed for a variety of values of transport properties (including the inertial relaxation time). Figures 1 and 2 show profiles of advective fluxes (effectively, concentrations) and diffusive fluxes at an instant of time, t, as a function of distance, x, from the origin of a semi-infinite, one-dimensional space. Both figures show results computed for t equal to 0.2 year, advective velocity, v, equal to 1 meter/year, and the coefficient of hydrodynamic dispersion, D, equal to 0.2 meter^2/year. In each case, the inner boundary condition was a constant total (advective plus dispersive) flux, J_0 mole/(meter^2 year), at the origin. In Figure 1, the ordinate is the dimensionless ratio of the advective flux, vC, where C is the concentration of solute in mole/meter^3, to the boundary flux, J_0. In Figure 2, the ordinate is the dimensionless ratio of the dispersive flux, j, to the boundary flux, J_0. Note that the dispersive flux is composed of two terms: the usual "Fickian" flux plus the inertial term. Graphs of inertial solutions are shown for two values of the relaxation time, τ, and their corresponding values of propagation velocity, v_w; corresponding solutions of the inertia-less transport equation ($\tau = 0$) are also shown for comparison.

PROPERTIES OF THE SOLUTIONS

Several properties of the solutions obtained, as illustrated in Figures 1 and 2, are interesting in the context of the nuclear waste disposal problem:

1. The leading edge of a solute front propagates at a finite velocity dependent on the relaxation time, the dispersion coefficient, and the mean advective velocity of fluid flow. In the limit as the relaxation time goes to zero, the

velocity of propagation increases without bound and the wave solution approaches the diffusion solution. Conversely, as the relaxation time increases in an advective system, the velocity of propagation approaches the advective velocity, and the wave solution approaches a distortionless translation of the initial material anomaly.

2. The leading edge of a solute front is truncated by a step function which decreases in magnitude exponentially with time.

3. Inertial effects are most evident near the leading edge of the solute front, but as time becomes large relative to the time of arrival of the leading edge at a given location the wave solution approaches the diffusion solution; i.e., the mass flux becomes "Fickian".

ESTIMATION OF THE RELAXATION TIME

Estimation of values of the relaxation time, τ, has proven to be very difficult. Chu and Sposito [11] have discussed the physical meaning of τ and have indicated how τ can be estimated from the appropriate experimental data. However, such data are very scarce; indeed, no experiments directed specifically toward estimating τ have been reported. Chu and Sposito [11], using other data, estimated a value of the order of 10^4 seconds (3×10^{-4} year) for sandy soils. This value is two to three orders of magnitude lower than the values used in the calculations shown in Figures 1 and 2, and it is clear that the inertial effect would be insignificant at this level. However, due to the nature of the data used by Chu and Sposito [11], their estimate is not precise and is more likely too low than too high.

A need for further research exists in two areas: (1) acquisition of data from appropriately designed experiments to more precisely define the magnitude of the inertial effect in engineered barrier materials, and (2) further development of the fundamental concepts underlying the theory of solute transport in porous media.

SUMMARY

In summary, it is evident that model calculations of breakthrough curves of radionuclides transported through engineered barriers could be erroneous near the leading edge if the inertial effect were sufficiently large. Conversely, theoretical interpretation of the results of laboratory measurements or of field monitoring experiments would be adversely affected by an incorrect choice of transport model. Because of a lack of data needed for precise evaluation of the inertial parameter, τ, considerable uncertainty exists about the significance of inertial effects in engineered barriers. However, it is clear that inclusion of inertial effects in mass transport provides, in principle, an improved physical basis for the evaluation of barrier performance.

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FIGURES

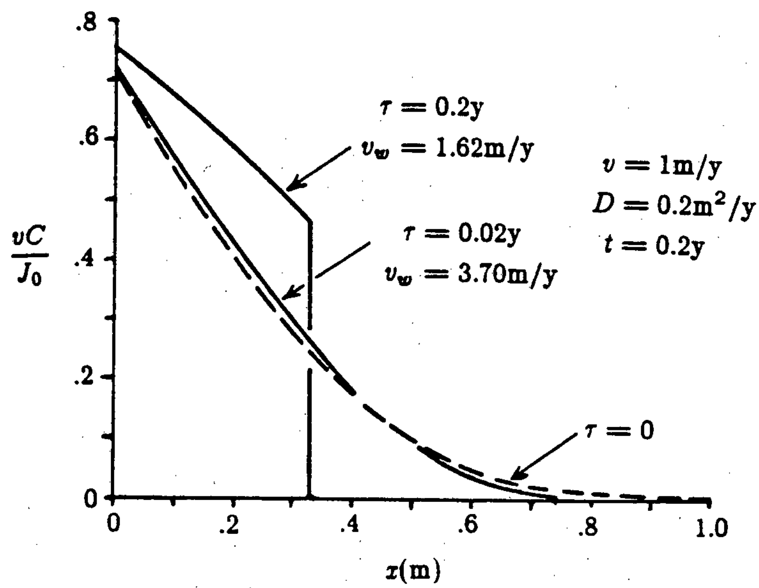

Fig. 1. Advective flux for flux boundary condition.

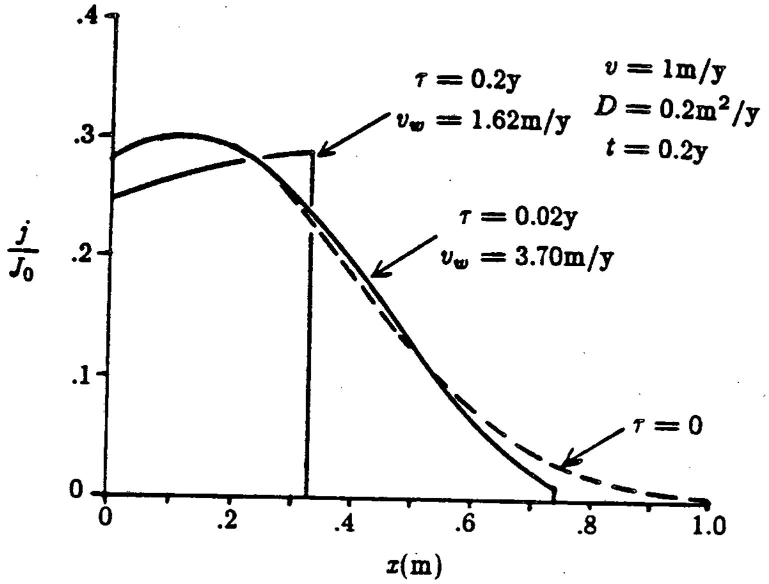

Fig. 2. Diffusive flux for flux boundary condition.

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